## UK Patent Application (19) GB (11) 2 042 562

- (21) Application No 8003483
- (22) Date of filing 1 Feb 1980
- (30) Priority data
- (31) 1104/79 8793/79
- (32) 5 Feb 1979 28 Sep 1979
- (33) Switzerland (CH)
- (43) Application published
  - 24 Sep 1980
- (51) INT CL<sup>3</sup> C08K 54/15 C07D 307/88 C08K 5/34
- (52) Domestic classification
  C3K 210 211 294 297 298
  CZ
  C2C 1474 200 213 220
  226 22Y 253 25Y 304 305
  30Y 351 352 366 368 386
  387 401 40Y 43X 491 635
  638 645 648 658 65X 668
  66X 699 761 767 806 80Y
  816 AA TU
  C3W 100 203 205 217
  218 221 317
- (56) Documents cited GB 1532478
- (58) Field of search C3K
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- (54) Improvements in or relating to stabilising polymers
- (57) The present invention relates to a process for stabilising organic polymeric materials comprising incorporating therein a benzofuran(2)-one compound or indolin(2)one compound containing at least two benzofuran(2)one or indolin(2)one nuclei.

## **SPECIFICATION**

## Improvements in or relating to organic compounds

5 The present invention relates to a process for stabilizing organic polymeric materials employing benzofuranone or indolinone compounds as stabilisers.

Accordingly, the present invention provides a process for stabilising organic polymeric materials comprising incorporating therein a benzofuran(2)one or indolin(2)one compound contain at least two benzofuran(2)one or indolin(2)one nuclei, respectively.

Preferred benzofuran(2) one compounds and indolin(2) one compounds for use in the process of the present invention are bis-benzofuran(2) one or bis-indolin(2) one compounds in which the 3-position of the first benzofuran(2) one or indolin(2) one nucleus is bound directly to the 3- or 7-position of the second benzofuran(2) one or indolin(2) one nucleus, respectively, or the 5-, 6- or 7-position of the first benzofuran(2) one or indolin(2) one nucleus is bound directly

to the same position of the second nucleus, and benzofuran(2)one or indolin(2)one compounds in which the 3-, 5-, 6-, or 7-position of the benzofuran(2)one or indolin(2)one nucleus is attached to the same position of 1 to 5 further such nuclei through a 2 to 6 valent bridge member.

Preferred directly bound bis-benzofuran(2)ones and bis-indolin(2)ones are those of formula l<sub>a</sub>,

in which X is -O- or -NR<sub>10a</sub> either, R<sub>a</sub> is hydrogen or (aa/1)

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and each R<sub>1a</sub>, independently, is hydrogen; C<sub>1-22</sub> - alkyl; C<sub>5-5</sub>cycloalkyl; C<sub>1-5</sub>alkyl-C<sub>5-6</sub>cycloalkyl; phenyl; phenyl substituted by a total of up to three substituents selected from the group consisting of C<sub>1-12</sub>alkyl (up to three of these with max. 18 carbon atoms in the combined alkyl substituents), hydroxyl (max. of two of these), C<sub>1-12</sub>alkoxy, C<sub>1-18</sub>acyloxy, chlorine and nitro (max. of one of each of these); a group of formula (a/4), (a/5) or (a/6)

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$$\frac{CR_{2}}{n} = \frac{CR_{8}}{C-NR_{8}} \qquad (a/5)$$

65 or, R, together with R, is (a/3)

$$R_{3a}$$
 $R_{3a}$ 
 $R_{3a}$ 
 $R_{3a}$ 
 $R_{3a}$ 

or, R<sub>a</sub> is hydrogen and

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$$R_{1a}$$
 is  $(a/7)$ 
 $R_{4a}$ 
 $C=0$ 
 $R_{3a}$ 
 $R_{2a}$ 
 $R_{1a}$ 
 $R_{1a}$ 

with the proviso that when  $R_{12}$  is (a/7) X is  $-O_{-}$ ,

 $R_{1ax}$  is phenyl or phenyl substituted by a total of up to three substituents selected from the group consisting of  $C_{1-12}$  alkyl (max. three of these with a total of up to 18 carbon atoms in the combined alkyl substituents), hydroxyl (max. two of these),  $C_{1-12}$  alkoxy,  $C_{1-18}$  acyloxy, chlorine and nitro (max. one of each of these),

and  $R_{2a}$  to  $R_{5a}$ , independently, is hydrogen;  $C_{1-12}$  alkyl;

max. two of R₃ to R₅ are:

90 C<sub>5-6</sub>cycloalkyl; C<sub>1-5</sub>alkyl-C<sub>5-6</sub>cycloalkyl; hydroxyl; C<sub>1-22</sub>alkoxy; phenoxy optionally substituted by up to two C<sub>1-12</sub>alkyl groups with a total of up to 16 carbon atoms in the combined alkyl substituents; C<sub>1-18</sub> - acyloxy; phenylcarbonyloxy; chlorine; max. one of 95 R<sub>2</sub> to R<sub>5</sub> is:-

phenyl- $C_{1-9}$ alkyl or phenylthio in which the phenylnucleus is optionally substituted by up to three substituents selected from  $C_{1-12}$ alkyl, hydroxyl, and  $R_{15}CO-O-$ ; phenyl optionally substituted by up to two  $C_{1-12}$ alkyl groups with a total of up to 16 carbon atoms in the combined substituents; nitro;

O 
$$\parallel$$
 $-C-R_{11}$  (b/2);
105  $-CH_2S-R_{12}$  (b/3) as  $R_3$ 
 $-CH(C_6H_5)CO-O-R_7$  (b/4) as  $R_3$  (a/4) or (a/5) as  $R_3$  or  $R_5$  with the proviso that when  $R_{11}$  in (b/2) is other than hydrogen such (b/2) group is adjacent a hydroxyl group,

or, when  $R_a$  is hydrogen,  $R_{1a}$  is other than (a/7) and X is -O-,  $R_{3a}$  is  $(E_3)$  or  $R_5$  is  $(E_5)$ 

 $R_7$ , is  $C_{1-18}$  alkyl; alkyl-O-alkylene with a total no. of 120 up to 18 carbon atoms; alkyl-S-alkylene with a total no. of up to 18 carbon atoms; di- $C_{1-4}$  - alkylamino  $C_{1-8}$  alkyl;  $C_{5-1}$  cycloalkyl; or phenyl optionally substituted by up to 3  $C_{1-12}$  alkyl groups with a total no. of up to 18 carbon atoms in the com- 125 bined substituents,

either, each R<sub>8</sub>, independently, is hydrogen;  $C_{1-18}$  alkyl;  $C_{5-6}$  cycloalkyl;  $C_{1-5}$  alkyl- $C_{5-5}$  cycloalkyl; phenyl optionally substituted by up to two  $C_{1-12}$  alkyl groups with max. 16 carbon atoms in the combined substituents;

-CH₂CH₂OH (d/1); -CH₂CH₂OC₁-₁€alkyl (d/2); or O

5 -CH<sub>2</sub>CH<sub>2</sub>-O-C-R<sub>11</sub> (d/3)

or, both R<sub>a</sub> together with the nitrogen form piperidine or morpholine,

R, has one of the significances of R,

R<sub>92</sub> is hydrogen, C<sub>1-18</sub>alkyl, (d/1), (d/2) or (d/3),

0 R<sub>10a</sub> is hydrogen,  $C_{1-16}$ alkyl,  $C_{5-6}$ cycloalkyl,  $C_{1-5}$ alkyl- $C_{5-6}$ cycloalkyl or phenyl optionally substituted by up to two  $C_{1-12}$ alkyl groups with max. 16 carbon atoms in the combined substituents, or benzyl,

15  $R_{11}$  is hydrogen,  $C_{1-22}$ alkyl,  $C_{5-7}$ cycloalkyl, phenyl $C_{1-6}$ alkyl or phenyl optionally substituted by up to two  $C_{1-12}$ alkyl groups with max. 16 carbon atoms in the combined substituents,

R<sub>12</sub> is C<sub>1-18</sub>alkyl, 2-hydroxyethyl, phenyl or

20 (C,\_9)alkylphenyl,

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 $R_{15}$  is  $C_{1-22}$ alkyl or phenyl, and n is 0, 1 or 2,

and the molecule contains only two benzofuran(2)one or indolin(2)one nuclei, whereby the substituents on the two benzofuran(2)one or indolin(2)one nuclei are the same or different, preferably they are the same.

Of the directly bound benzofuran(2)ones and indolin(2)ones, the benzofuran(2)one compounds are preferred.

Preferred poly-benzofuran(2)ones or indolin(2)ones linked by a bridging group are those in which the bridging group is bound to the 3-, 5- or 7-position of the benzofuran(2)one or indolin(2)one 35 nucleus.

When the bridging group is attached to the 3-position of the benzofuran(2)one or indolin(2)one nucleus, such group may be bound via a single or double bond.

40 Preferred bridged benzofuran(2) one or indolin(2) one compounds are those of formula l<sub>b</sub>,

in which -X - is as defined above,

 $R_b$ ,  $R_{1b}$ ,  $R_{2b}$ ,  $R_{2b}$ ,  $R_{4b}$  and  $R_{5b}$  correspond to the significances  $R_a$ ,  $R_{1a}$ ,  $R_{2a}$ ,  $R_{3a}$ ,  $R_{4a}$  and  $R_{5a}$  above with the exception that the molecule is free from groups of formulae (aa/1), (a/3), (a/7), (E<sub>3</sub>) and (E<sub>5</sub>) and

either  $R_{1b}$ , or  $R_b$  and  $R_{1b}$  together, or  $R_{3b}$ , or  $R_{5b}$  is bound to one or more further corresponding benzofuran(2)one or indolin(2)one nuclei through a polyvalent bridge member.

Preferred groups in place of Rib are:-

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$$\begin{array}{c|c}
0 & & \\
\hline
0 & -c & \\
\hline
0 & & \\
c_{1-4}alkyl
\end{array}$$
(e/7a)

in which the free valencies are attached to groups E,

A is a 2 to 6 valent saturated alkylene which optionally contains sulphur, oxygen, nitrogen or cyclohexylene bridges or is a 2- or 3-valent benzene radical or when both Z's are -O-, À is also (e/16)

whereby when A is a 3-, 4-, 5- or 6-valent radical the further valencies are bound to OH, -NHR<sub>10</sub> or

groups, with the proviso that any free valencies on nitrogen in A itself are attached to

w is from 1 to 6, each Z, independently, is -0- or  $-NR_{10}$ ,

 $R_{10}$  has one of the significances of  $R_{10a}$  above or  $R_{10}$  together with the N-atom signifies

D is a direct bond or -O-, -S-,

$$R_{13}$$
  
|-SO<sub>2</sub>-> C = O or -C-  
|  
| R<sub>13</sub>

in which each  $R_{13}$ , independently, is hydrogen,  $C_{1-16}$  alkyl (preferably  $C_{1-4}$ -alkyl) with the proviso that when both  $R_{13}$  are alkyl the combined groups contain

max. 16 carbon atoms, phenyl, (a/4) or (a/5);

n is as defined above,

m is 2 to 10, and

s is 0 or 1 to 12.

5 Preferred groups in place of R<sub>b</sub> and R<sub>1b</sub> together

$$= \overset{R_{16}}{c} - \overset{O}{c} - z - \lambda + \left( z - \overset{O}{c} - \overset{R_{16}}{c} \right) \qquad (e/9)$$

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(e/13)

15 in which the free valencies are attached to groups E12

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and A, Z, w and  $R_{10}$  are as defined above, with the exception that on A the further free valencies are

attached to -OH, -NHR<sub>10</sub> or -Z-CR<sub>16</sub>=E<sub>1a</sub>, and any free valencies on nitrogen, in A itself are

attached to -C-CR<sub>16</sub>=E<sub>1a</sub>,

p is 0 or 1 to 10, and R<sub>16</sub> is hydrogen or methyl.

Preferred groups in place of R<sub>3b</sub> are:-

in which R<sub>13</sub> is as defined above,

or (e/1), or (e/4) in which the free valencies are attached to groups E36

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and A, w, Z and R<sub>10</sub> are as defined above, with the exception that on A the further free valencies are attached to -OH, -NHR, or

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and any free valencies on nitrogen in A itself are attached to

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Preferably when R<sub>30</sub> is a bridging group bound to one or more Eponuclei, X is -O- in all cases.

Preferred groups in place of Rsb are:-

in which R<sub>13</sub> is as defined above, or

(e/1), or (e/4) in which the free valencies are attached to groups Esh

and A, w, Z and  $R_{10}$  are as defined above, with the exception that the further free valencies on A are attached to -OH, -NHR10 or

and any N-free valencies in A itself are attached to

Preferably when Rsb is a bridging group bound to E<sub>sb</sub>, X is -O- in all cases.

Examples of 2-valent -Z-A-Z- groups are:-

-Z (CH<sub>2</sub>)--Z- where n<sub>1</sub> is 2 to 12,

90 -O(CH<sub>2</sub>CH<sub>2</sub>O)<sub>1,2 or 3</sub>CH<sub>2</sub>CH<sub>2</sub>O--OCH2CH2SCH2CH2O-, -OCH2CH2S-SCH2CH2O-,

 $R_{10}$  $-N-C_qH_{2q}O-$  where q is 2 or 3,

-OCH2CH2N CH2CH2O-,

Examples of 3 valent -Z-A-Z- groups are:-

$$-HN-(CH_2)_3-N(CH_2CH_2O-)_2$$
,  $N(CH_2CHO-)_3$ ,  $-Z-CH_2C-(CH_2-Z-)_2$ ,

C1-6alkyl

N(CH2CH2O-)3

130 -HN-(CH<sub>2</sub>)-2013-N-CH<sub>2</sub>CH<sub>2</sub>-O-, HN-(CH<sub>2</sub>CH<sub>2</sub>-O-)<sub>2</sub>,

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Examples of 4 valent -Z-A-Z- groups are:-( $-OCH_2$ ),-C,

An Example of a 5-valent -Z-A-Z- groups is:-

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-O-CH-(-CH)<sub>4</sub>-CH<sub>2</sub>-O- and  $[(-O-CH_2)_3-C-CH_2]_{\frac{1}{2}}$  O A is preferably 2-, 3- or 4-valent with the following -Z-A-Z- groups being most preferred: -Z-(CH<sub>2</sub>)<sub>n</sub>-Z- with  $n_1 = 2$  to 6 or 10

especially those in which A is alkylene.

Most preferred Z-A-Z groups are:-

 $C(CH_2-O-)_4$  and  $-O-(CH_2)_n$  O— with  $n_1 = 2$  to 6 or 10 s is preferably 0 to 10 more preferably, 0 to 8. m is preferably 2 to 6.

p is preferably 2 or 3, more preferably 3.

When  $R_{1b}$  is a bridge member carrying one or more benzofuran(2)one or indolin(2)one nuclei it is preferably  $R_{1b}$ ' where  $R_{1b}$ ' is (e/1), (e/4), (e/6), (e/7), (e/7a), (e/7b) or (e/8) more preferably (e/7) or (e/8).

When R<sub>1b</sub> and R<sub>b</sub> together form a bridge member carrying one or more benzofuran(2)one or indolin(2)one nuclei they preferably form (e/9) or (e/13).

When  $R_{3o}$  is a bridge member carrying one or more benzofuran(2)one groups it is preferably  $R_{3o}$ ,

$$R_{13}$$
 |  $R_{13}$  |  $R_{13}$  |  $R_{13}$  |  $R_{13}$  |  $R_{13}$ 

(e/1) or (e/4), where each R<sub>13</sub>', independently, is hyd-55 rogen, C<sub>1-4</sub>alkyl or (a/4) in which R<sub>7</sub> is hydrogen, C<sub>1-18</sub>alkyl (preferably methyl) with the proviso that when one R<sub>13</sub>' is (a/4) the other R<sub>13</sub>' is other than (a/4) preferably methyl. More preferably R<sub>30</sub> as a bridging member carrying one or more benzofuran(2) one

60 nuclei, is R<sub>10</sub>" where R<sub>10</sub>" is

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(e/1) or (e/4), especially (e/1).

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When  $R_{sb}$  is a bridge member carrying one or more benzofuran(2)one groups it is preferably  $R_{sb}$  where  $R_{sb}$  is

where  $R_{13}$ ' is as defined above or (e/1). More preferably  $R_{5b}$  as a bridge member is  $R_{5b}$ ", where  $R_{5b}$ " is -S- or  $-CH_2-$ .

In the compounds of formula  $l_b$  the substituents on each benzofuran(2)one or indolin(2)one nucleus are the same or different, preferably they are the same.

Of the benzofuran(2)one or indolin(2)one compounds having a bridge member bound to further such nuclei, the benzofuran(2)one compounds are preferred.

 $R_a$  is preferably  $R_a'$ , where  $R_a'$  is hydrogen or (aa/1) with (aa/1) being most preferred. In (aa/1) preferably  $R_{\tau a}$  to  $R_{5a}$  have the preferred significances as stated herein.

In (a/7),  $R_{1ax}$ ,  $R_{2a}$ ,  $R_{2a}$ ,  $R_{3a}$  and  $R_{4a}$  preferably have the preferred significances stated herein.

In (a/3) preferably  $R_{2a}$  to  $R_{5a}$  have the preferred significances stated herein.

R<sub>1a</sub> is preferably R<sub>1a</sub>', where R<sub>1a</sub>' is (a/7) or R<sub>1</sub>',

95 where R<sub>1</sub>' is hydrogen, C<sub>1-18</sub>alkyl, phenyl optionally substituted by one or two C<sub>1-8</sub>alkyl groups and/or a hydroxyl group; (a/4) or (a/5). More preferably R<sub>1a</sub> is R<sub>1</sub>", where R<sub>1</sub>" is C<sub>1-18</sub>alkyl or phenyl optionally substituted by one or two (C<sub>1-18</sub>)alkyl groups and/or a hydroxyl group. Most preferably R<sub>1a</sub> is R<sub>1</sub>", where R<sub>1</sub>" is phenyl optionally substituted by C<sub>1-4</sub>alkyl, with unsubstituted phenyl being most preferred.

R<sub>1ax</sub> is preferably R<sub>1</sub>", most preferably phenyl optionally substituted by C<sub>1-4</sub>alkyl, with unsubstituted phenyl being most preferred.

 $R_{2a}$  and  $R_{2b}$  are preferably  $R_2'$ , where  $R_2'$  is hydrogen or  $C_{1-a}$  alkyl, more preferably  $R_2''$ , where  $R_2''$  is hydrogen or methyl, with hydrogen being especially preferred.

In E<sub>3</sub> preferably R<sub>a</sub>, R<sub>1a</sub>, R<sub>2a</sub>, R<sub>4a</sub> and R<sub>5a</sub> have the preferred significances stated herewith.

 $R_{3a}$  is preferably  $R_{3a}$ , where  $R_{3a}$  is  $E_3$  or  $R_3$ , where  $R_3$  is hydrogen or  $C_{1-9}$  (preferably  $C_{1-5}$ ) alkyl. Most preferably  $R_{3a}$  is  $R_3$ .

115 R<sub>4a</sub> and R<sub>4b</sub> are preferably R<sub>4</sub>', where R<sub>4</sub>' is hydrogen or C<sub>1-4</sub>alkyl, with hydrogen being especially preferred.

In  $E_5$  preferably  $R_4$  and  $R_{14}$  to  $R_{14}$  have the preferred significances stated herein.

T<sub>5a</sub> is preferably  $R_{5a}$ , where  $R_{5a}$  is  $E_5$  or  $R_5$ , where  $R_5$  is hydrogen or  $C_{1-a}$  (preferably  $C_{1-5}$ ) alkyl. More preferably  $R_{5a}$  is  $R_5$ .

 $R_b$  is preferably  $R_b$ ' where  $R_b$ ' is hydrogen or together with  $R_{b}$  is (e/9) or (e/13).

125  $R_{15}$  is preferably  $R_{15x}$ , where  $R_{15x}$  is either  $R_1$ , more preferably  $R_1$ " especially phenyl, or  $R_{15}$ , especially with the preferred A groups in (e/1) and with s as 1 to 8, preferably 4 in (e/8).

 $R_{3b}$  is preferably  $R_{3b}$ , where  $R_{3b}$  is hydrogen, 130  $C_{1-3}$  alkyl or  $R_{3b}$ , more preferably hydrogen, methyl

or R<sub>36</sub>".

 $R_{sb}$  is preferably  $R_{sbx}$ , where  $R_{sbx}$  is hydrogen,  $C_{1-8}$ alkyl or  $R_{50}$ , more preferably hydrogen or  $R_{50}$ .

5 X is preferably -O- or -NR<sub>102</sub>', where R<sub>10a</sub>' is hydrogen, (C<sub>1-12</sub>)alkyl, phenyl or benzyl, more preferably, hydrogen, (C1-4)alkyl or phenyl. Most preferably X is -O-.

 $R_{10}$  is preferably  $R_{10}$ , where  $R_{10}$  is hydrogen, 10 (C1-12)alkyl or phenyl, more preferably hydrogen or (C1-4)alkyl, especially hydrogen or methyl.

 $R_7$  in (a/4) and (b/4) is preferably  $R_7$  where  $R_7$  is hydrogen, C1-18alkyl, phenyl optionally substituted by up to two C₂-12alkyl groups with max. 16 carbon 15 atoms in the combined substituents. More preferably  $R_7$  is  $R_7$ ", where  $R_7$ " is  $C_{1-18}$  alkyl, phenyl or C1-12alkylphenyl. Most preferably R7 is C1-18alkyl, especially Ca-18alkyl.

Each R<sub>a</sub>, independently, is preferably R<sub>a</sub>', where R<sub>a</sub>' 20 is hydrogen, C<sub>1-18</sub> alkyl or both R<sub>8</sub>'s together form morpholine or piperidine. More preferably each R<sub>8</sub>, independently is hydrogen or C1-18 alkyl. Preferred alkyl groups as  $R_8$  are  $C_{1-12}$ -, preferably  $C_{1-8}$ -, most preferably C1-4alkyl.

R<sub>9</sub> is preferably R<sub>9</sub>', where R<sub>9</sub>' is hydrogen, C<sub>1-8</sub>alkyl or (d/1). More preferably R<sub>9</sub> is hydrogen or C1-8 alkyl. The preferred alkyl as R9 contains 1 to 4 carbon atoms.

R<sub>92</sub> is preferably R<sub>92</sub>', where R<sub>92</sub>' is hydrogen or 30 C<sub>1-8</sub>alkyl. Any alkyl as R<sub>92</sub> preferably contains 1 to 1 to 4 carbon atoms.

R<sub>11</sub> is preferably R<sub>11</sub>, where R<sub>11</sub> is hydrogen,  $C_{1-18}$ alkyl or phenyl.  $R_{11}$  in (b/2) is preferably phenyl. Any alkyl as R<sub>11</sub> preferably contain 1 to 17 carbon 35 atoms.

 $R_{12}$  is preferably  $R_{12}$ , where  $R_{12}$  is  $C_{1-12}$  alkyl, phenyl or 4-(alkyl C<sub>1-9</sub>)phenyl.

R<sub>16</sub> is preferably hydrogen.

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n in (a/4) or (a/5) as  $R_1$  or  $R_5$  is preferably 1.

n in (a/4) or (a/5) as  $R_3$  is preferably 2.

Preferred compounds of formula la, are those in which X is  $-O-R_a$  is  $R_a'$ ,  $R_{1a}$  is  $R_{1a}'$ ,  $R_{2a}$  is  $R_2'$ , preferably  $R_2''$ ,  $R_{3a}$  is  $R_{3a}'$ ,  $R_{4a}$  is  $R_4'$  and  $R_{5a}$  is  $R_{5a}'$  with the proviso that the molecule contains two and only two

45 directly bound benzofuran(2) one nuclei.

More preferred compounds of formula la are those in which Ra is (aa/1), both Ria's are Ri", more preferably R,", especially phenyl, R2a is hydrogen, R1a is  $R_3$ ', preferably hydrogen,  $R_{4a}$  is hydrogen,  $R_{5a}$  is  $R_5$ ', 50 preferably hydrogen and X is -O- and each benzofuran(2)one nucleus is identical.

When R<sub>30</sub> is a bridge member carrying further benzofuran(2)one nuclei preferably R<sub>2</sub> and R<sub>4</sub> are both hydrogen and Rsb is hydrogen or C1-4alkyl, especially 55 hydrogen.

When Rss is a bridge member carrying further benzofuran(2)one nuclei preferably R<sub>36</sub> is hydrogen or

Preferred compounds of formula I<sub>b</sub> are those in 60 which  $R_b$  is  $R_b'$ ,  $R_{1b}$  is  $R_{1ba}$ ,  $R_{2b}$  is  $R_2'$ , preferably  $R_2''$ ,  $R_{3b}$  is  $R_{3bx}$ ,  $R_{4b}$  is  $R_{4}$ ,  $R_{5b}$  is  $R_{5bx}$  and X is -O-, and preferably each benzofuran(2) one nucleus is identical, with the proviso that only one bridge member bearing one or more benzofuran(2) one nuclei is pre-65 sent in the molecule.

More preferred compounds of formula lb are those in which  $R_b$  is  $R_b'$ ,  $R_{1b}$  is  $R_{1bx}$ ,  $R_{2b}$  is hydrogen,  $R_{3b}$  is hydrogen, methyl or  $R_{3b}$ ",  $R_{4b}$  is hydrogen,  $R_{5b}$  is hydrogen or R<sub>sb</sub>", X is -O- and each benzofuran(2) one 70 nucleus is identical.

The directly bound his benzofuran(2)one or indolin(2) one compounds are either known or may be prepared in accordance with known methods from available starting materials.

75 The bridged benzofuran(2)one and indolin(2)one compounds as defined above are new and also form part of the present invention.

The bridged benzofuran(2)ones and indolin(2)ones of the invention may be prepared by con-80 ventional methods. For example, by condensing a polyfunctional bridge member with appropriately substituted benzofuran(2)ones or indolin(2)ones or by condensation and ring closure reactions of for example, hydroxy substituted bridged benzenes, 85 and by interconversion reactions.

For example compounds of formula l<sub>b</sub> in which one of  $R_1$ ,  $R_3$  or  $R_5$  is (e/1) or (e/4), may be prepared by reacting a corresponding monomeric benzofuran(2)one compound or indolin(2)one compound in 90 which one of R₁, R₃ and R₅ is

or a functional derivative thereof with a compound H–Z–A–(Z–H), compound or with a di- or triaminobenzene or with a di- or tri-hydroxybenzene or for (e/4) with piperazine in known manner. Preferred functional derivatives are acid chlorides and lower 100 alkylesters.

Compounds of formula I<sub>b</sub> in which R<sub>1</sub> is (e/5) or (e/6) may be prepared by reacting a corresponding monomeric benzofuran(2) one or indolin(2) one compound in which R and R, are both hydrogen with a 105 compound of formula

following by catalytic hydration in accordance with 110 known methods.

Similarly, the compounds of formula lo in which R and R, together form (e/9), (e/12) or (e/13) may be prepared by reacting the corresponding monomeric benzofuran(2)one or indolin(2)one compound where 115 R and R, are both hydrogen with the corresponding

aldehyde of each of the bridge members. The compounds of formula lo in which R, is (e/7), (e/7a), (e/7b) or (e/8) can be prepared by reacting the

corresponding monomeric benzofuran(2) one or 120 indolin(2) one compound in which

$$R_1$$
 is  $C_{1-4}^{OH}$   $C_{1-4}^{OH}$ 

with ноос -{О,- соон

125 or a functional derivative thereof, with HOOC-(CH<sub>2</sub>)<sub>5</sub>-COOH for (e/8) or a functional derivative thereof, with COCI2 for (e/7a), and 130 with [O]<sub>0.1</sub> PCI<sub>3</sub> for (e/7b),

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in accordance with known methods. Preferred functional derivatives are acid chlorides and lower alkyl esters.

The compounds of formula  $l_b$ , in which  $R_{3b}$  is

O 
$$(R_{13})_2$$
  
 $| | |$   
 $-O-E_{3b}$ ,  $-S-E_{3b}$ ,  $-SO_2E_{3b}$ ,  $-C-E_{3b}$ ,  $-C-E_{5b}$ , or  $R_{5b}$  is
$$(R_{13})_2$$

-S-E<sub>3b</sub> or -C-E<sub>5b</sub> by reacting a compound of formula III or IV

15 
$$R_{2b} \xrightarrow{R_{2b}} R_{4b} \xrightarrow{R_{5b}} R_{4b} \xrightarrow{R_{3b}} R_{3b}$$

$$R_{2b} \xrightarrow{R_{4b}} R_{4b} \xrightarrow{R_{4b}} R_{3b} \qquad \text{IV for } R_{5b}$$
20

OH |

with a compound of formula  $R_{1a}$ —CH—C—OH in a 25 1:2 molar ratio, where Q is  $-O_-$ ,  $-S_1-$ ,  $-SO_2-$ , C=O

$$(R_{13})_2$$
  
|  
or -C-  
 $(R_{13})_2$ 

.Q, is -S- or -C-, and

 $R_1$  is optionally substituted phenyl as given for  $R_{1b}$  above, by known methods.

The compounds of formula  $l_b$  in which R and R, are both hydrogen may be prepared by reacting a compound of formula V

in which  $R_{2b}$  to  $R_{5b}$  are as defined above and one of  $R_{3b}$  or  $R_5$  is a bridge member as defined above linked to a further such phenolic compound, and with the exception that in any group (a/4) or (b/4),  $R_7$  is hydrogen, and  $R_{3b}$  and  $R_{5b}$  are other than (a/5),

G is a secondary amine group or halogen, with an ionic cyanide compound, hydrolysing the product thereof, followed by a ring closure condensation.

G is preferably –N(C<sub>1-4</sub>)alkyl or morpholine, especially –N(CH<sub>3</sub>)<sub>2</sub>. Any halogen as G is preferably chlorine or bromine, especially chlorine. Suitable ionic cyanide compounds are alkali- or alkaline earth cyanides, preferably sodium- or potassium cyanide.
 Each of the reaction steps may be carried out in accordance with known methods for such reactions.

The end product may be esterified or etherified to obtain compounds where R, is other than hydrogen. Furthermore, the methylene group in the 3-position

may be reacted further to obtain compounds where R, is other than hydrogen.

The compounds of formula III, IV and V are either known or may be prepared by known methods from available starting materials. The same applies for the other starting materials.

The directly bound bis-benzofuran(2)ones, bis-indolin(2)one and bridged benzofuran(2)ones or indolin(2)ones as defined above (hereinafter referred to as compounds K) may be incorporated into the polymeric material to be stabilized before, during, or after polymerization.

The amount of compound K incorporated may vary according to the material to be stabilized and the ultimate use to which it is to be put. Suitable amounts are from 0.01 to 5% preferably from 0.05 to 1%, based on the weight of the materials to be stabilized. The organic polymeric materials to be stabilized may be natural or synthetic polymeric materials. Examples of such materials include rubber, polyolefins, especially polyethylene, polypropylene, ethylene, propylene copolymers, polybutylene, polystyrene, chlorinated polyethylene, PVC, polyester, polycarbonate, polymethylmethacrylate, polyphenyleneoxide, polyamides such as nylon, polyurethanes, polypropyleneoxide, phenolformaldehyde resins, epoxy resins, polyacrylonitrile and corresponding copolymers such as acrylonitrile butadiene styrene (ABS) terpolymers.

The process of the present invention is preferably
employed to stabilise polypropylene, polyethylene,
ethylene/propylene copolymers, PVC, polyesters,
polyamides, polyurethanes, polyacrylonitrile, ABS
terpolymers, terpolymers of acrylic ester, styrene
and acrylonitrile, copolymers of styrene and
acrylonitrile, styrene/butadiene copolymers,
polybutylene and polystyrene. The most preferred
organic polymeric materials are polypropylene,
polyethylene especially HDPE, ethylene/propylene
copolymers and ABS.

The incorporation of the compounds K in the material to be stabilized is effected in accordance with know methods. Preferred methods are those in which the compounds K are incorporated in the polymeric material by melt blending the stabiliser and the additives in conventional equipments such as Banbury mixers, extruders etc. Polypropylene and polyethylene granulates on powders are advantageously employed, whereby the compounds of formula I are admixed with said powders and then extruded etc and worked into the films, foils, bands threads etc.

The process of the present invention may be carried out by incorporating a compound K alone or together with other additives e.g. further stabilisers etc.

The preferred process according to the present invention comprises incorporating a compound K and either (i) a stabiliser of the sterically hindered phenol type,

or (ii) a sulphur-containing or phosphorous containing stabiliser,

or (i) and (ii), into the polymeric material.

The ratio of stabiliser (i) or (ii) to the compounds K incorporated in the polymeric material is suitably 5:1 to 1:5, preferably 2:1 to 1:1. The ratio of combined (other) stabilisers to compounds of formula l is suitably 15:1 to 1:5, preferably 6:1 to 1:3. Preferably, when only stabilisers (i) are employed with the compounds of formula l<sub>c</sub> the ratio of compounds (i) to 130 those of formula l<sub>c</sub> is 3:1 to 1:1.

Examples of sterically hindered phenols are:β - (4 - hydroxy - 3,5 - ditert. - butylphenyl) - propionicacidstearyl - ester, tetrakis methylene - 3(3',5' ditert. - butyl - 4 - hydroxyphenyl) - propionate] -5 methane, 1,3,3 - tris - (2 - methyl - 4 - hydroxy - 5 tert. - butylphenyl) - butane, 1,3,5 - tris(4 - tert. - butyl -3 - hydroxy - 2,6 - dimethylbenzyl) - 1,3,5 - triazine -2,4,6 - (1H,3H,5H) - trione, bis - (4 - tert. - butyl - 3 hydroxy - 2,6 - dimethylbenzyl) - dithiolterephtha-10 late, tris(3,5 - ditert. - butyl - 4 - hydroxybenzylisocyanurate, triester of 3,5 - di - tert. - butyl - 4 hydroxyhydrocinnamic acid with 1,3,5 - tris - (2 hydroxyethyl) - s - triazin - 2,4,6 - (1H,3H,5H) - trione, bis [3,3 - bis - 4' - hydroxy - 3 - tert. - butylphenyl) -15 butaneacid - glycolester, 1,3,5 - trimethyl - 2,4,6 - tris - (3,5 - ditert. - butyl - 4 - hydroxybenzyl) - benzene, 2,2' - methylene bis (4 - methyl - 6 - tert. - butylphenyl) terephthalate, 4,4 - methylene - bis - (2,6 ditert. - butylphenol), 4,4' - butylidene - bis - (6 - tert. -20 butyl - meta - cresol), 4,4 - thio - bis(2 - tert. - butyl - 5 - methyl - phenol), 2,2' - methyl - bis(4 - methyl - 6 tert. - butylphenol.

Examples of sulphur containing stabilisers are disteary! - thiodipropionate, dilaury!thiodipropionate, 25 tetrakis(methylene - 3 - hexylthiopropionate) - methane, tetrakis (methylene - 3 - dodecy! - thiopropionate) - methane and dioctadecy!disulphide.

Examples of phosphorous containing compounds are trinonyl - phenylphosphite, 4,9 - distearyl - 30 3,5,8,10 - tetraoxadiphosphaspiroundecane, tris - (2,4 - ditert. - butylphenyl)phosphite and tetrakis(2,4 - ditert.butylphenyl) - 4,4' - diphenylene diphosphonite.

In addition to the above further stabilisers, U.V.
35 absorbers as described in DOS 2 606 358 e.g. 2 - (2' - hydroxyphenyl) - benztriazole, 2 - hydroxybenzophenone, 1,3 - bis(2 - hydroxybenzoyl)benzene, salicylates, cinnamic acid esters, hydroxybenzoic acid esters, sterically hindered amines and oxylic
40 acid diamides. Suitable such compounds are described in DOS 2 606 358.

Metal deactivators for example N,N' - dibenzoylhydrazide, N - benzoyl - N' - salicyloylhydrazide, N,N' - distearylhydrazide, N,N' - bis - [3 - (3,5 - ditert. - butyl - 4 - hydroxyphenyl) - propionyl] - hydrazide, N,N' - bis - salicyloylhydrazide, oxalylbis - (benzylidenehydrazide), N,N' - bis(3 - methoxy - 2 - naphthoyl - )hydrazide, N,N' - di - α - phenoxy - butyloxy (isophthalyl - dihydrazide) may also be incorporated into the polymeric material.

Additional conventional additives may also be employed for example, flame retardants, antistatic agents etc.

Furthermore, an optical brightener may be incor-55 porated in the polymer to be stabilised and so that the distribution of the additives which are intimately admixed with said optical brightener may be ascertained by fluorescence intensity measurements.

The present invention also provides master
batches of polymeric organic materials containing 5
to 90%, preferably 20 to 60%, more preferably
20-30% of a compound K. Such master batches may
then be admixed with unstabilised polymeric material. It is to be appreciated that such master batches
may also contain additional additives such as those

stated above.

Polymeric materials containing a compound K are primarily stabilised against degradation during processing. When, of course, other additives such as antioxidants, e.g. above phenols, and U.V. absorbers are also employed together with the compounds K the polymeric material has an enhanced long term stability against thermal- and photoxidative degradation.

The following examples further serve to illustrate the invention. In the examples all parts are by weight, and all temperatures are in degrees Centigrade.

Example 1

2.54 parts of the compound of formula

(prepared in accordance with known methods) and 1 part mandelic acid are heated to 200°C for 16 hours. Afterwards the reaction mixture is separated by column chromatography (silicagel, ether/petroliumether 1:2). Crystals having a melting point range of 185-187°C, corresponding to the formula of Compound No. 5 of the Table are obtained. Example 2

95 78.9 parts of the compound of formula

100

85

are dissolved in 450 parts diethyleneglycolmonomethylether. 39 Parts potassium cyanide and 6 parts potassium iodide are added thereto. At a temperature of 80°C, 63 parts of water are added dropwise. 105 The temperature is raised to 130° and the mixture is stirred for 16 hours at this temperature. After cooling to room temperature, 1000 parts ice water are added. After carefully acidifying with hydrochloric acid, a precipitate is formed which is dissolved in 400 110 parts ether. The organic phase is separated, washed with water, dehydrated over MgSO4 and evaporated. The residue is added to toluene, heated to the boil for approximately 1 hour whereupon water of condensation separates out. After evaporating the sol-115 vent and recrystallizing from methanol a colourless crystalline product of formula

is obtained. A mixture of 19.36 parts of the compound, 5.36 parts terephthalic aldehyde, 0.24 parts piperidine benzoate and 100 parts toluene are 125 heated for 15 hours at reflux temperature. After evaporation of the solvent the product is recrystallized from acetone. The so-obtained crystals are washed with a small amount of ice-cold ether and dried. A yellow powder having a melting point range 130 of 241-242°C corresponding to the formula of Com-

pound No. 9, is obtained.

Example 3

2, 0 Parts pentaerythritol - tetra - [3 - (4 - hydrox-yphenyl) - propionate] and 2, 1 parts mandelic acid are heated together to 180°C for 23 hours. After cooling, the reaction mixture is separated by column chromatography (silicagel, eluent 9:1 toluene/acetone). The so-obtained product has a melting point range of 90-95°C, and corresponds to the formula of Compound No. 10, Compound Nos. 1, 2 and 4 are made in an analogous manner, starting with the corresponding bis phenol compound and reacting the same with mandelic acid. Example 4

A solution of 2, 0 parts terephthalic acid dichloride in 40 parts toluene is added slowly at room temperature to a mixture of 5, 1 parts of the starting material used in Example 1, 100 parts toluene and 2, 1 parts triethylamine. A white precipitate is obtained. The mixture is stirred for some hours at room temperature followed by stirring at 80°C for 2 hours. The precipitate is filtered off and the clear solution is evaporated. A white crystalline product, melting point 245-246°C, (recrystallized from acetone/petroliumether) of the formula of Compound No. 6 is obtained. Compound No. 7 can be prepared in analogous manner.

Example 5

1, 60 Parts of Compound No. 9 are catalytically
 dehydrogenated at room temperature under normal pressure. As solvent 20 parts of glacial acetic acid is used, as catalyst 0.2 parts palladium on barium sulfate. After removal of the catalyst and the solvent, the residue is taken up with ether. The etherified solution is shaken with a Na-bicarbonate solution and then with water, followed by dehydrating over MgSO<sub>4</sub> and then evaporated. The product, having a melting point range of 258-259°C (petroliumether), corresponds to the formula of Compound No. 8.
 Compound No. 3 of the Table is prepared in accor-

dance with known methods.

Example A

A mixture of 1200 parts of a commercially available unstabilized polypropylene (Profax 6501), 0.6 105 parts calciumstearate, 0.6 parts tetrakis - methylene - 3(3',5' - di - tert. - butyl - 4 - hydroxyphenyl) - propionate) - methane and 0.6 parts of the Compound No. 8 of the Table are shaken together for 10 minutes and extruded at 120 revs/min with temperatures of 110 150, 240, 260, and 200° in the different heating areas of the extruder to form a strand which is granulated after passing through a water bath. The granulate is extruded and granulated a further 9 times, each time a part is taken to measure the Melt Flow Index (MFI 115 according to ASTM D 1238 L, 230°; 216 kg) which serves as a measure of the thermomechanical oxidative degradation of a polymer. A control without Compound 8 of the Table is also extruded in like manner and tested. In comparison, the polymer con-120 taining Compound No. 8 of the Table exhibits a greatly improved melt stability during continuous extrusion. The other compounds of the Table may be employed in like manner.

Example B

125 100 Parts unstabilized HD-polyethylene powder (Phillips Type) are stabilized with 0.02 parts Compound No. 2 of the Table and 0.01 parts tetrakis - [methylene - 3(3',5' - di - tert. - butyl - 4' - hydroxyphenyl) - propionate) - methane. The powder is sub-130 jected to a modified MFI Test at 230°/0.325 kg on a

Davenport-MFI apparatus. The powder is pushed into a heated steel cylinder and a 325 g weight is placed thereon. The polymer which is pressed out is cut off at 60 second intervals. The amount is calculated in terms of g/10 min. The stronger the crosslinking of the polymer owing to insufficient stabilization, the lower the MFI value. After 5 to 15 minutes a constant value is obtained. The other compounds of the Tables may be used in analogous manner.

1.0 Part octylstearate, 1.5 parts Ba-Cd stabilizer (powder-forming), 1 part of Compound No. 3 of the Table and 0.5 parts of a commercially available arylalkylphosphate are mixed with 100 parts com-15 mercially available dispersion PVC (k-value-60) in a Fluid Mixer (Papenmeier Type TEHK8) until the temperature has risen to 110°. The homogeneous mixture is rolled on rollers heated to 180° for 1 minute and then pressed into plates (thickness 1 20 mm) at 200° for 1.5 min. at 2 atm. and 1.5 minutes at 20 atm. The test or plates are put into an air circulating drying cabinet at 180°C for 30 minutes. A comparison sample which contained 2.5 parts Ba-Cd stabilizer instead of Compound No. 4 and 1.5 parts of 25 the Ba-Cd stabilizer was also treated in the same manner. This sample undergoes discolouration even at the beginning of the heat treatment and is markedly more discoloured after the 30 minutes than the sample containing Compound No. 4 of Table 1. 30 Example D

300 Parts ABS powder (Fa. Marbon AOE 30/075) are dissolved in 2200 parts chloroform and the solution is dropped into 8000 parts methanol whereupon the ABS is precipitated. After filtration the polymer 35 which is now free from stabilizer is treated in vacuo overnight to remove all the solvent. 100 parts of the so-treated ABS powder is dissolved in chloroform and 0.2 parts Compound No. 2 of the Table are added thereto and the whole is stirred under nit-40 rogen atmosphere for 15 minutes. The solution is drawn into a film with a 1 mm doctor blade onto a glass plate and is left for the solvent to evaporate-off whereby the film shrinks to 150  $\mu$  thickness and is freed from the rest of the solvent overnight at room 45 temperature in vacuo. The film is then stoved in an air-circulating oven at 95°. By repeated IRmeasurement to  $\Delta \epsilon = 0.4$  at 1715 cm<sup>-1</sup> the ageing resistance is checked. The samples containing the benzofuranone compound have longer resistance 50 than the control samples which contain no stabilizer.

Example E

100 Parts granulated polyethyleneterephthalate are ground to a rough powder and dried overnight at 100° in a vacuum drying cabinet. 1.0 Part of Com-55 pound No. 2 of the Table is added and the mixture is homogenised, then granulated in an extruder, spun into fibres at 280°, stretched (120 den/14) and twisted. The fibres are wound on to white cards and exposed to the light in an Atlas Weatherometer for 60 24 hour intervals. In comparison to a non-stabilized control, the sample containing Compound No. 41 has less tendency to yellow during the exposure to light and can be left in the Weatherometer for a substantially longer period of time in order to reach the

65 same decrease in the tensile strength (50%).

Example F

1000 Parts 20% styrene-butadiene rubber emulsion are added with stirring to a hydrochloric acid 5% sodium chloride solution whereupon the rubber coagulates. Stirring is continued for 1 hour at pH 3,5. After filtration the coagulate is repeatedly washed and dried to a constant weight at room temperature in a vacuum cabinet.

25 Parts of this rubber are heated under nitrogen
75 atmosphere to 125° in Brabender plastographs and mixed with 0.25 parts Compound No. 2 of Table 1 for 10 minutes and subsequently pressed to (0.5 mm thick) plates at 125°. The plates were put into an Atlas Weatherometer for 24 hour intervals together with 80 samples containing no stabilizer. In comparison to the latter samples, the stabilized samples exhibited significantly better resistance to light.

Example G

49.5 Parts Compound No. 2 of the Table, 49.5 parts tetrakis - [methylene - 3 - (3',5' - di - tert. - butyl - 4' - hydroxyphenyl) - propionate] - methane, 1 part calcium stearate and 0.02 parts (7 - [24 - naphthol(1,2d)triazol - 2 - yl] - 3 - phenylcumarine (optical brightener) are heated to 140°. The mixture melts with stirring and the melt is poured into a flat dish and ground after cooling. The product obtained melts at 70-75°C.

0.5 Parts of the ground melt are mixed in a plastic bag by repeated shaking with 1000 parts unstabilized 95 HDPE powder (Ziegler Type, MFI 190/z = 0.7). 43 Parts of the powder mixture are heated to 220° in a Brabender Plasti-Corder PLV 151 extruder at 50 revs/min. until there is a sharp drop in the torque indicating degradation (to cross-linking decreases). The 100 test sample has good stability.

When different concentrations of the above melt product are mixed with polyethylene or polypropylene powder and extruded into a strand which are subsequently ground, the fluorescence intensity can be measured to assess the distribution of additives in the polymer mixture.

CLAIMS

- A process for stabilising organic polymeric materials comprising incorporating therein a ben-110 zofuran(2)one compound or indolin(2)one compound containing at least two benzofuran(2)one or indolin(2)one nuclei.
- A process according to Claim 1, in which either a bis benzofuran(2)one or bis indolin(2)one com115 pound in which the 3-position of the first benzofuran(2)one or indolin(2)one nucleus is bound directly to the 3- or 7-position of the second benzofuran(2)one or indolin(2)one nucleus, respectively, or the 5-, 6- or 7-position of the first benzofuran(2)one
   120 or indolin(2)one nucleus is bound directly to the
- same position of the second nucleus or a benzofuran(2) one or indolin(2) one compound in which the 3-, 5-, 6- or 7-position of the benzofuran(2) one or indolin(2) one nucleus is attached to the same position of 1 to 5 further such nuclei through a 2 to 6
- valent bridge member is incorporated in the polymeric material.
- 3. A process according to Claim 2, in which the bis benzofuran(2)one or indolin(2)one compound is 130 of formula I<sub>a</sub>,

in which X is -O- or -NR<sub>102</sub> either, R<sub>2</sub> is hydrogen or (aa/1)

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10  $R_{4a} \longrightarrow R_{3a} \times C=0$  (aa/1)

and each R<sub>12</sub>, independently, is hydrogen;
 C<sub>1-22</sub>alkyl; C<sub>5-6</sub>cycloalkyl; C<sub>1-5</sub>alkyl-C<sub>5-6</sub>cycloalkyl;
 phenyl; phenyl substituted by a total of up to three substituents selected from the group consisting of C<sub>1-12</sub>alkyl (up to three of these with max. 18 carbon
 atoms in the combined alkyl substituents), hydroxyl (max. of two of these), C<sub>1-12</sub>alkoxy, C<sub>1-18</sub>acyloxy, chlorine and nitro (max. of one of each of these); a group of formula (a/4), (a/5) or (a/6)

25 
$$\frac{\circ}{-(CR_2 + n - C - CR_7)}$$

$$\frac{\circ}{-(CR_2 + n - C - CR_8)}$$

or, Ra together with Rta is (a/3)

or, Ra is hydrogen and

35

40

$$R_{1a} = 1s = (a/7)$$
 $R_{1a} = 1s = (a/7)$ 
 $R_{1a} = 1s = (a/7)$ 

45 with the proviso that when  $R_{1a}$  is (a/7) X is -0-,

R<sub>1ax</sub> is phenyl or phenyl substituted by a total of up to three substituents selected from the group consisting of C<sub>1-12</sub>alkyl (max. three of these with a total of up to 18 carbon atoms in the combined alkyl substituents), hydroxyl (max. two of these), C<sub>1-12</sub>alkoxy, C<sub>1-18</sub>acyloxy, chlorine and nitro(max. one of each of these),

and  $R_{2a}$  to  $R_{5a}$ , independently, is hydrogen;  $C_{1-12}$  alkyl;

55 max. two of R<sub>2</sub> to R<sub>3</sub> are:

 $C_{5-6}$ cycloalkyl;  $C_{1-5}$ alkyl- $C_{5-6}$ cycloalkyl; hydroxyl;  $C_{1-22}$ alkoxy; phenoxy optionally substituted by up to two  $C_{1-12}$ alkyl groups with a total of up to 16 carbon atoms in the combined alkyl substituents;

60 C<sub>1-10</sub>acyloxy; phenylcarbonyloxy; chlorine; max. one of R<sub>2</sub> to R<sub>3</sub> is:-

phenyl- $C_{1-9}$ alkyl or phenylthio in which the phenyl nucleus is optionally substituted by up to three substituents selected from  $C_{1-12}$ alkyl, hydroxyl, and

65 R<sub>15</sub>CO-O-; phenyl optionally substituted by up to

two  $C_{1-12}$ alkyl groups with a total of up to 16 carbon atoms in the combined substituents; nitro;

O
||
|70 -C-R<sub>11</sub> (b/2);
-CH<sub>2</sub>S-R<sub>12</sub> (b/3) as R<sub>3</sub>
-CH(C<sub>6</sub>H<sub>5</sub>)CO-O-R<sub>7</sub> (b/4) as R<sub>3</sub>
(a/4) or (a/5) as R<sub>3</sub> or 8<sub>5</sub>

with the proviso that when  $R_{\tau\tau}$  in (b/2) is other than 75 hydrogen such (b/2) group is adjacent a hydroxyl group,

or, when  $R_a$  is hydrogen,  $R_{1a}$  is other than (a/7) and X is -0-,  $R_{3a}$  is (E<sub>3</sub>) or  $R_s$  is (E<sub>5</sub>)

80

 $R_7$ , is hydrogen  $C_{1-10}$ alkyl; alkyl—O-alkylene with a total no. of up to 18 carbon atoms; alkyl—S-alkylene with a total no. of up to 18 carbon atoms; di- $C_{1-4}$ alkylamino $C_{1-6}$ alkyl;  $C_{5-7}$ cycloalkyl; or phenyl optionally substituted by up to 3  $C_{1-12}$ alkyl groups with a total no. of up to 18 carbon atoms in the combined substitutuents,

either, each  $R_8$ , independently, is hydrogen;  $C_{1-18}$ alkyl;  $C_{5-6}$ cycloalkyl;  $C_{1-5}$ alkyl- $C_{5-6}$ cycloalkyl; phenyl optionally substituted by up to two  $C_{1-12}$ alkyl groups with max. 16 carbon atoms in the combined substituents;

-CH₂CH₂OH (d/1); -CH₂CH₂OC₁-₁8alkyl (d/2); or O

100  $-CH_2CH_2-O-C-R_{11}$  (d/3) or, both  $R_\theta$  together with the nitrogen form piperidine or morpholine,

 $R_9$  has one of the significances of  $R_8$ ,  $R_{9a}$  is hydrogen,  $C_{1-18}$ alkyl, (d/1), (d/2) or (d/3),

05  $R_{10a}$  is hydrogen,  $C_{1-18}$ alkyl,  $C_{5-6}$ cycloalkyl,  $C_{1-5}$ alkyl- $C_{5-6}$ cycloalkyl or phenyl optionally substituted by up to two  $C_{1-12}$ alkyl groups with max. 16 carbon atoms in the combined substituents, or benzyl

110 R<sub>11</sub> is hydrogen, C<sub>1-22</sub>alkyl, C<sub>5-7</sub>cycloalkyl, phenylC<sub>1-6</sub>alkyl or phenyl optionally substituted by up to two C<sub>1-12</sub>alkyl groups with max. 16 carbon atoms in the combined substituents,

 $R_{12}$  is  $C_{1-18}$ alkyl, 2 - hydroxyethyl, phenyl or 115  $(C_{1-9})$ alkylphenyl,

 $R_{15}$  is  $C_{1-22}$ alkyl or phenyl, and

 $R_{15}$  is  $C_{1-22}$  alkyl or phenyl, and n is 0, 1 or 2,

and the molecule contains only two benzofuran(2)one or indolin(2)one nuclei, whereby the sub-120 stituents on the two benzofuran(2)one or indolin(2)one nuclei are the same or different, and the bridged benzofuran(2)one or indolin(2)one compound is of formula l<sub>b</sub>

in which -X is as defined above, 130  $R_b$ ,  $R_{1b}$ ,  $R_{2b}$ ,  $R_{3b}$ ,  $R_{4b}$  and  $R_{5b}$  correspond to the sig15

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nificances  $R_a$ ,  $R_{1a}$ ,  $R_{2a}$ ,  $R_{3a}$ ,  $R_{4a}$  and  $R_{5a}$  above with the exception that the molecule is free from groups of formulae (aa/1), (a/3), (a/7), (E<sub>3</sub>) and (E<sub>5</sub>) and

either R<sub>1b</sub>, or R<sub>b</sub> and R<sub>1b</sub> together, or R<sub>3b</sub>, or R<sub>5b</sub> is 5 bound to one or more further corresponding benzofuran(2)one or indolin(2)one nuclei through a polyvalent bridge member.

4. A process according to Claim 3, in which any bridging group carrying further benzofuran(2)one or 10 indolin(2)one nuclei as R<sub>1b</sub> is

$$C_{1-4}^{\circ}alky1$$

$$C_{1-4}alky1$$

$$C_{1-4}alky1$$

$$\begin{array}{c|c}
0 & & \\
\hline
C_{1-4}alky1 & & \\
\end{array}$$
(e/7:.)

$$C_{1-4}^{O} = C_{1-4}^{O} =$$

in which the free valencies are attached to groups E

$$45 \qquad \qquad \underset{R_{3b}}{\overset{R_{4b}}{\underset{R_{2b}}{\bigvee}}} \overset{\text{if Sb}}{\underset{R_{2b}}{\bigvee}} x_{C^{*0}} \qquad \qquad \epsilon_{1}$$

A is a 2 to 6 valent saturated alkylene which optionally contains sulphur, oxygen, nitrogen or cyclohexylene bridges or is a 2- or 3-valent benzene radical or when both Z's are -O-, A is also (e/16)

whereby when A is a 3-, 4-, 5- or 6-valent radical the further valencies are bound to OH, –NHR<sub>10</sub> or

groups, with the proviso that any free valencies on nitrogen in A itself are attached to

w is from 1 to 6,

70

75

85

100

120

130

(e/5)

each Z, independently, is -O- or -NR10,

 $R_{10}$  has one of the significances of  $R_{10a}$  above or  $R_{10}$  together with the N-atom signifies

D is a direct bond or -O-, -S-,

$$R_{13}$$
 | -SO<sub>2</sub>-> C=0 or -C- |  $R_{13}$ 

in which each  $R_{13}$ , independently, is hydrogen,  $C_{1-16}$  alkyl with the proviso that when both  $R_{13}$  are alkyl the combined groups contain max. 16 carbon atoms, phenyl, (a/4) or (a/5);

n is as defined above,

m is 2 to 10, and

s is 0 or 1 to 12,

any bridging group carrying further benzofuran(2)one or indolin(2)one nuclei as  $R_{b}$  and  $R_{1b}$  together is

in which the free valencies are attached to groups E12

and A, Z, w and  $R_{10}$  are as defined above, with the exceptions that on A the further free valencies are attached to –OH, –NHR<sub>10</sub> or

and any free valencies on nitrogen, in A itself are attached to

p is 0 or 1 to 10, and

R<sub>16</sub> is hydrogen or methyl, any bridging group carrying further benzofuran(2)one or indolin(2)one nuclei as R<sub>36</sub> is

125 in which R<sub>13</sub> is as defined above, or (e/1), or (e/4) in which the free valencies are attached to groups E<sub>36</sub>

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60

| |-Z-C-(CH₂)<sub>n</sub>-E₃<sub>b</sub>

and any free valencies on nitrogen in A itself are attached to

10

and any bridging group carrying further benzofuran(2) one or indolin(2) one nuclei as R<sub>50</sub> is

15

in which  $R_{13}$  is as defined above, or 20 (e/1), or (e/4) in which the free valencies are attached

to groups E₅ь

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and A, w, Z and  $R_{10}$  are as defined above, with the exception that the further free valencies on A are attached to -OH,  $-NHR_{10}$  or

30

and any N-free valencies in A itself are attached to

35

with the proviso that only one of  $R_{1b}$ ,  $R_b$  and  $R_{1b}$  together,  $R_{3b}$  or  $R_{5b}$  is a bridge member carrying further benzofuran(2)one or indolin(2)one nuclei, and the substituents on the nuclei are the same or different.

- 5. A process according to Claim 4, in which when  $R_{3b}$  or  $R_{5b}$  is a bridging group, X is -0—in all cases.
- 6. A process according to Claim 3, in which X is 45 0.
  - 7. A process according to Claim 4, in which R<sub>a</sub> is hydrogen or (aa/1),

 $R_{1a}$  is  $R_{1a}$ , where  $R_{1a}$  is (a/7) or  $R_{1}$ , where  $R_{1}$  is hydrogen,  $C_{1-18}$ alkyl, phenyl optionally substituted by one or two  $C_{1-18}$ alkyl groups and/or a hydroxyl group; (a/4) or (a/5).

 $R_{1ax}$  is  $C_{1-1a}$  alkyl or phenyl optionally substituted by one or two  $C_{1-1a}$  alkyl groups and/or a hydroxyl group, each of  $R_{2a}$  and  $R_{2b}$  is hydrogen or  $C_{1-4}$  alkyl,

 $R_{3a}$  is  $R_{3a}$ , where  $R_{3a}$  is  $E_3$  or  $R_3$ , where  $R_3$  is hydrogen or  $C_{1-9}$  alkyl, each of  $R_{4a}$  and  $R_{4b}$  hydrogen or  $C_{1-4}$  alkyl,

 $R_{5a}$  is  $R_{5a}$ , where  $R_{5a}$  is  $E_{5}$  or  $R_{5}$ , where  $R_{5}$  is hydrogen or  $C_{1-a}$ alkyl,

R<sub>b</sub> is hydrogen or together with R<sub>1b</sub> is (e/9) or (e/13) R<sub>1b</sub> is R<sub>1bx</sub> where R<sub>1bx</sub> is either R<sub>1</sub>' or is (e/1), (e/4), (e/6), (e/7), (e/7a), (e/7b) or (e/8),

Rab is Rabe, where Rabe is hydrogen or Canalkyl or

65

 $-S-E_{3b}$ ,  $-C-E_{b}$ , (e/1) or (e/4)

(R13')2

in which each  $R_{13}$ , independently, is hydrogen,

 $(C_{1-4})$ alkyl or (a/4) in which  $R_7$  is hydrogen or  $(C_{1-18})$ alkyl, with the proviso that when one of  $R_{13}$  is 70 (a/4) the other  $R_{13}$  is other than (a/4),

Rs is Rsbr, where Rsbx is hydrogen, C1-8alkyl or

(R<sub>13</sub>') | |-S-E<sub>5b</sub>, -C-E<sub>5b</sub> or (e/1), and

 $R_{10}$  is hydrogen,  $C_{1-2}$ alkyl or phenyl.

8. A process according to any one of Claims 3 to 7, in which X is +O-.

9. A process according to Claim 7, in which any R<sub>1</sub>' is C<sub>1-18</sub>alkyl or phenyl optionally substituted by one or two C<sub>1-8</sub>alkyl groups and/or a hydroxyl group.

10. A process according to Claim 7, in which X is -0— and in each benzofuran(2) one nucleus is identical with the exception of the case where  $R_{1a}$  is (a/7) where  $R_{1a}$  in the second nucleus is  $R_{1ax}$ .

11. A process according to any one of Claims 3 to 10, in which  $R_{1a}$  is (aa/1).

12. A process according to any one of Claims 3 to 10, in which  $R_{1a}$  is (a/7).

13. A process according to any one of Claims 4 to 10, in which  $R_b$  is hydrogen.

14. a process according to any one of Claims 1 to 13, in which the polymeric material is polypropylene, polyethylene, ethylene/propylene copolymers, PVC, polyesters, polyamides, polyurethanes, polyacrylonitrile, ABS terpolymers, terpolymers of acrylic ester, styrene and acrylonitrile, copolymers of styrene and acrylonitrile, styrene/butadiene

copolymers, polybutylene or polystyrene.

15. A process according to Claim 14, in which the 100 polymeric material is polypropylene.

16. A process according to Claim 14, in which the polymeric material is polyethylene or an ethylene/propylene copolymer.

17. A process according to Claim 16, in which the105 polymeric material is high density (HD) polyethylene.

18. A process according to any one of Claims 1 to 17, in which from 0.01 to 5%, based on the weight of the polymeric material, of the benzofuran(2)one or indolin(2)one compound is incorporated in the polymeric material to be stabilised.

19. A process according to any one of Claims 1 to17, in which the benzofuran(2)one or indolin(2)one compound is incorporated into the polymeric mater-115 ial by melt blending.

20. A process according to any one of Claims 1 to 17 or 19, in which 5 to 90% of the benzofuran(2) one or indolin(2) one compound is incorporated in the polymeric material to form a stabilized master batch.

120 21. A process according to any one of Claims 1 to 19, comprising incorporating the benzofuran(2) one or indolin(2) one compound together with

either (i) a stabiliser of the sterically hindered phenol type

125 or (ii) a sulphur-containing or phosphorouscontaining stabiliser or (i) and (ii),

into the polymeric material to be stabilised.

- 22. A process for stabilising organic polymeric material substantially as hereinbefore described with reference to any one of Examples A to G.
- 23. Polymeric organic material whenever stabil-5 ised by a process according to any one of Claims 1 to22.
  - 24. A benzofuran(2)one or indolin(2)one compound, in which the 3-, 5-, 6- or 7-position of the benzofuran(2)one or indolin(2)one nucleus is
- 10 attached to the same position of 1 to 5 further such nuclei through a 2 to 6 valent bridge member.
  - 25. A compound of formula  $l_{\text{b}}$ , as defined in Claim 3.
- 26. A compound of formula  $l_{\text{b}}$ , as defined in 15 Claim 4.
  - 27. A compound of formula  $I_b$ , in which X is -0-.
  - 28. Compound Nos. 1, 2, 4 or 6 to 10 of the Examples.

Printed for Her Majesty's Stationery Office by The Tweeddale Press Ltd., Berwick-upon-Tweed, 1980.
Published at the Patent Office, 25 Southampton Buildings, London, WC2A 1AY, from which copies may be obtained.